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AIRBORNE TRITIUM AT LAWRENCE LIVERMORE LABORATORY SITE BOUNDARY

C. L. Lindeken
W. J. Silver
A. J. Toy
J. H. White

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CORRELATION BETWEEN PREDICTED AND OBSERVED LEVELS OF AIRBORNE
TRITIUM AT LAWRENCE LIVERMORE LABORATORY SITE BOUNDARY*

C. L. Lindeken, W. J. Silver, A. J. Toy and J. H. White

Lawrence Livermore Laboratory, University of California
Livermore, California 94550

ABSTRACT

At the Lawrence Livermore Laboratory, a computer code based on the Gaussian plume model is used to estimate radiation doses from routine or accidental release of airborne radioactive material. Routine releases of tritium have been used as a test of the overall uncertainty associated with these estimates. The ratio of concentration to release rate at distances from the two principal release points to each of six site boundary sampling locations has been calculated using local meteorological data. The concentration of airborne tritiated water vapor is continuously measured at the six sampling stations as part of the Laboratory's environmental monitoring program. Comparison of predicted with observed annual tritiated water concentrations in 1978 showed an average ratio of 2.6 with a range of from 0.97 to 5.8.

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INTRODUCTION

At the Lawrence Livermore Laboratory (LLL), a computer code based on the Gaussian plume model is used to estimate radiation doses from routine or accidental release of airborne materials.⁽¹⁾ Included in these estimates is the dose that could be received by a hypothetical person at a point on the site boundary closest to a release point, as well as the dose to the nearest off-site resident. These estimates are included in the Laboratory's annual environmental monitoring report.⁽²⁾ The need for validating the computer code has long been recognized; however, experiments specifically designed to test its predictive accuracy, such as those in which controlled quantities of radioactivity are released, are now environmentally unacceptable. As part of the LLL environmental monitoring program, tritiated water vapor is continuously measured at each of six air-monitoring stations on the site perimeter. As a test of the overall uncertainty associated with our dose estimates, we have compared code-predicted concentrations of tritium resulting from annual routine releases with those concentrations observed through field monitoring.

TRITIUM RELEASE POINTS

Low levels of tritium are routinely released to the atmosphere from Building 212, which houses the Insulating Core Transformer Accelerator (ICT), and from Building 331, the Tritium Research Facility (TRF). Figure 1, an LLL Site map, shows the location of these facilities and the distribution of the six monitoring stations.

The ICT is mainly used to generate 14 MeV neutrons by means of the ^3H (d, n) ^3He reaction. Ventilating air from two target chambers is exhausted through stacks 2-m above the roof. The TRF was constructed in two increments--each equipped with 30-m stacks. Air from all radioactive-material areas within the facility is exhausted through these stacks.

Many of the high-flux experiments involving tritium targets that were conducted at the ICT are now conducted in Building 292, The Rotating Target Neutron Source (RTNS-II), located in the northwest quadrant of LLL. Because of the tritium containment and recovery system in use at this facility, tritium releases to the atmosphere are negligible and Building 292 is not considered a tritium release point.

CONCENTRATION TO RELEASE RATE RATIOS

The ratio of concentration to release rate (χ/Q) at the distances from the two release points to each of the six site boundary sampling locations has been calculated using local meteorological data from an instrumented tower. This tower, located near the Laboratory's north boundary, is equipped with wind direction, wind speed and temperature sensors mounted at 10- and 40-m levels. From records of these data, wind speed, wind direction and atmospheric stability estimates were tabulated every 1/2 h over a calendar year. Variance in the horizontal wind direction was used to estimate Pasquill-Gifford stability categories based on the method described by Slade.⁽³⁾ Lateral and vertical standard deviations, σ_y and σ_z , are entered in the computer code as functions of these stability categories and the respective distances. The average annual χ/Q value was calculated for

the distance from each tritium release point to each monitoring station. All calculations are corrected for seasonal wind frequencies. Table 1 shows the direction of each of the site boundary sampling stations from the release points, the distances between them and the χ/Q values.

Effluent records show that 1100 Ci of tritium were released from the ICT during 1978. During the same period 4200 Ci were released from the TRF. Measurements made on both of the TRF stack effluents throughout the year, in which HT and HTO distributions are made with differential molecular sieves, showed that approximately half of the tritium released was in the form of tritiated water. No such analyses were performed on the ICT effluent so it was conservatively assumed that all the tritium was released in the form of tritiated water.

AIRBORNE TRITIATED WATER MEASUREMENTS

Water vapor is collected at each of the perimeter monitoring locations by drawing air at flow rates of about 0.5 l/m through silica gel absorbers. Flow rates are maintained by the use of critical orifices. Absorbers are changed at 2-week intervals.

In the laboratory the silica gel is transferred to a tared plastic bag, weighed and then well mixed by tumbling. A portion of this gel is then transferred to a tared freeze-dried jar. After weighing, the gel is freeze dried and the tritium content of the recovered water is determined by liquid-scintillation counting.

RESULTS AND DISCUSSION

Table 2 shows the predicted annual average concentration of tritiated water vapor at each monitoring location calculated from the 1978 releases from the respective facilities assuming a uniform release rate throughout the year. Table 3 shows monthly and annual average concentrations of tritiated water observed by environmental monitoring. Table 4 compares the calculated annual averages with those obtained from the monitoring data. The average ratio of predicted/observed concentration was 2.6 with a range from 0.97 to 5.8.

The assumption that all the tritium released by the ICT was in the form of HTO, although conservative for dose estimates, is probably the reason for the generally high predicted/observed concentration ratios. Plans call for routine measurements of the HT and HTO distribution in the ICT effluent using the same differential molecular sieve technique employed at the TRF. Concentrations measured at Location 12 are often more than twice as high as the average of all other stations due to its close proximity to the ICT and because it is typically downwind from this source, as shown by the wind rose in Figure 2. The influence of upwind buildings in reasonably close proximity to the monitoring stations at location 12 and 14 may also lower the observed concentrations through enhanced diffusion and cloud dispersal.

In balance, considering the uncertainties in the assumptions, the average agreement within a factor of three between predicted and observed concentrations of tritium is quite encouraging. The work to date illustrates how site boundary monitoring measurements and effluent release data may be applied in validation testing of atmospheric diffusion models.

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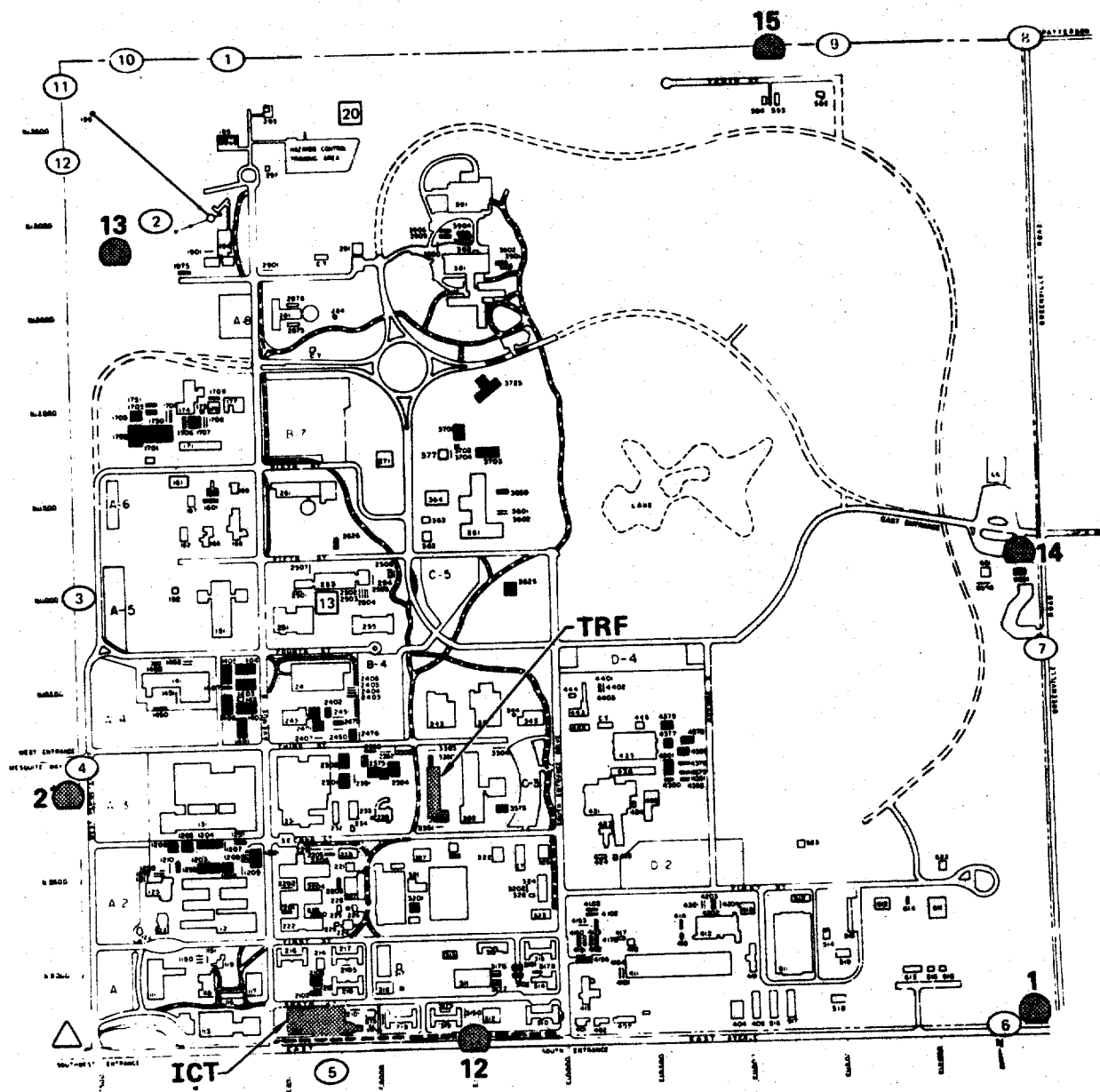


FIG. 1. Location of perimeter monitoring stations and tritium release points at LLL.

Table 1. Calculated ratios of concentration to release rate (X/Q) for tritium at LLL site boundary monitoring locations.

Sampling location	From ICT			From TRF		
	Direction of plume (a)	Distance to sampler, m	X/Q ₃ s/m ³	Direction of plume	Distance to sampler, m	X/Q ₃ s/m ³
1	E	1143	2.1E-6	ESE	1052	3.4E-7
2	NW	617	1.3E-6	W	594	7.3E-7
12	E	229	1.5E-5	S	377	6.8E-7
13	NNW	1234	4.4E-7	NW	983	2.7E-7
14	NE	1349	2.0E-7	ENE	1029	1.3E-6
15	NNE	1772	4.2E-7	NNE	1360	3.3E-7

(a) Direction of plume to reach sampling station

Table 2. Concentration of HTO at LLL Site Perimeter Sampler Stations Based on Diffusion Estimates

Sampling Station	ICT Effluent, (μCi/ml)	TRF Effluent, (μCi/ml)	Total, (μCi/ml)
1	7.4 E-11	2.3 E-11	9.7 E-11
2	4.6 E-11	4.9 E-11	9.5 E-11
12	5.3 E-10	4.6 E-11	5.8 E-10
13	1.5 E-11	1.8 E-11	3.4 E-11
14	7.1 E-11	8.7 E-11	1.6 E-10
15	1.5 E-11	2.2 E-11	3.7 E-11

TABLE 3. Tritium (HTO) concentrations in air at the LLL perimeter during 1978.

10⁻¹¹ μCi/ml

Location	Jan.	Feb.	Mar.	Apr.	May	Jun.	Jul.	Aug.	Sep.	Oct.	Nov.	Dec.	Average
Perimeter													
1	4.3	3.6	4.1	3.5	3.1	3.9	4.4	2.8	2.8	2.4	3.3	3.8	3.5
2	11.0	8.7	8.5	5.7	4.0	2.1	2.5	3.0	3.4	7.2	6.6	5.1	5.6
12	13.1	7.3	12.0	9.8	9.2	11.6	11.2	10.1	18.7	N.S.*	7.0	12.5	10.1
13	5.2	2.9	2.7	2.4	2.1	2.3	1.9	1.8	1.5	2.8	3.5	2.9	2.7
14	5.2	3.3	5.5	6.4	3.8	7.6	8.7	5.9	4.1	4.0	4.2	5.7	5.3
15	8.5	3.9	3.3	4.7	4.1	2.4	3.7	3.2	2.6	2.5	2.7	3.8	3.8

* No Sample

Table 4. Comparison of predicted and observed HTO concentrations at LLL Site Boundary

Sampling Station	$\mu\text{Ci/ml}$		
	Predicted	Observed	Predicted/Observed
1	9.7 E-11	3.5 E-11	2.77
2	9.5 E-11	5.6 E-11	1.70
12	5.8 E-10	1.0 E-10	5.80
13	3.4 E-11	2.7 E-11	1.26
14	1.6 E-10	5.3 E-11	3.02
15	3.7 E-11	3.8 E-11	0.97

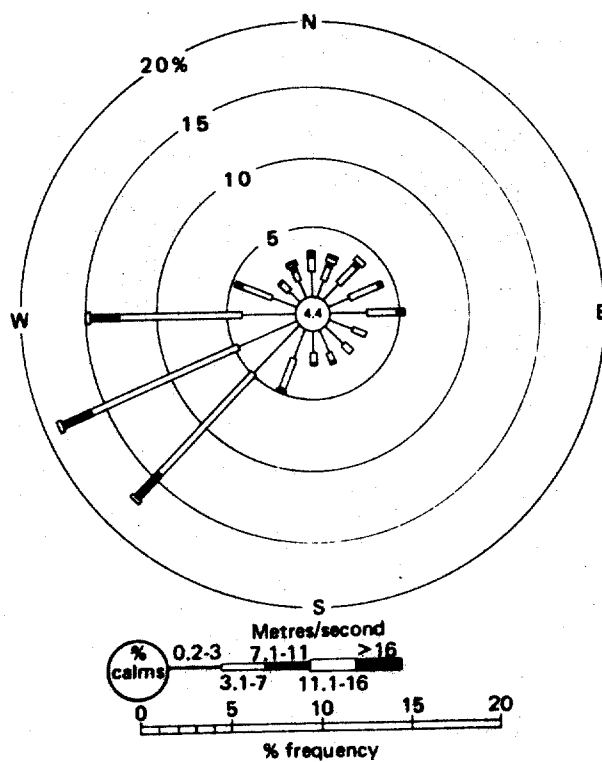


FIG. 2. Typical annual windrose for LLL.